

DISTRIBUTED CONTAINER FAILURE MODELS FOR THE DUST-MS COMPUTER CODE

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**Under Contract No. DE-AC02-98CH10886 with the
UNITED STATES DEPARTMENT OF ENERGY**

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ABSTRACT

Improvements to the DUST-MS computer code have been made that permit simulation of distributed container failure rates. The new models permit instant failure of all containers within a computational volume, uniform failure of these containers over time, or a normal distribution in container failures. Incorporation of a distributed failure model requires wasteform releases to be calculated using a convolution integral. In addition, the models permit a unique time of emplacement for each modeled container and allow a fraction of the containers to fail at emplacement. Implementation of these models, verification testing, and an example problem comparing releases from a wasteform with a two-species decay chain as a function of failure distribution are presented in the paper.

INTRODUCTION

Disposal of low-level radioactive (LLW) wastes requires a demonstration that environmental concentrations of radionuclides do not exceed regulatory limits chosen to ensure the protection of public health. This requires the quantitative assessment of the potential radiological impact of a LLW disposal facility on the surrounding environment. Evaluation of these impacts is accomplished through a performance assessment which includes estimates of the following processes for each radionuclide: (a) the rate of release from the disposal unit (i.e., the source term); (b) the transport from the disposal unit to the accessible environment; and (c) the conversion of the radionuclide concentration at the receptor site into an equivalent dose.

The objective of the DUST-MS (Disposal Unit Source Term – Multiple Species) computer model is to provide a tool that estimates the radionuclide release rate from the disposal facility, that is, the source term (1). In general, the source term is influenced by the radionuclide inventory and its origin (i.e., waste stream), the wasteforms and containers used to dispose of the inventory, and the physical processes that lead to release from the facility. DUST-MS may also be used to simulate transport through the unsaturated zone down to the aquifer. In addition, a recent improvement to DUST-MS includes a feature that creates an output file of mass flux at specified locations (1). Through selecting the proper location, (i.e. at the top of the aquifer), DUST-MS can be run a second time to simulate transport in the aquifer and the output file containing mass flux values from the first simulation (disposal facility and unsaturated zone) can be used as the inlet boundary condition for the second simulation (aquifer zone). This approach conserves mass between the two simulations.

The models selected to represent the four major processes (fluid flow, container degradation, wasteform leaching, and radionuclide transport) influencing release and transport have been incorporated into the computer code DUST-MS. Wasteform release is modeled through three release mechanisms:

- a) a surface rinse process in which radionuclides are released upon contact with the solution, partitioning between the wasteform and solution can be modeled;
- b) diffusion controlled release from the wasteform; and
- c) uniform release in which a fixed fraction of the inventory is released every year.

* This work was performed under the auspices of the U.S. Department of Energy.

All of these release mechanisms account for radioactive decay and ingrowth of the source. In addition, a check is performed to insure that releases do not cause concentrations to exceed a user-defined solubility limit. Transport is modeled using a one-dimensional finite-difference solution of the advection/dispersion equation. The model considers the physical/chemical processes of advection, diffusion, dispersion, radioactive decay and ingrowth, and external sources (wasteform release rates) and sinks.

Through support provided by Idaho National Engineering and Environmental Laboratory (INEEL), improvements have been made to the container failure model in DUST-MS. These include:

- a) Allowing a unique burial time for each container. In practice, a disposal site may be open for many years. Inventory values are reported at the time of disposal. The improved model permits a user to specify a problem start time (i.e., time at which waste was first disposed) and a disposal time for each container. This improves the accuracy for calculating releases radionuclides that have a half-life on the order of the operational time of the facility or less.
- b) Allowing time-distributed container failures. In previous versions of the model, container degradation was modeled through a unique container failure time. The value for this parameter should be selected based on the materials and expected environment. It was recognized that in using the one-dimensional DUST-MS code a single modeled container often represents a series of containers. In practice the failure time of each container in the series will be different. To accommodate this, DUST-MS was generalized to permit a distribution of container failures. The distribution will be specified using either a uniform failure rate or a Gaussian (normal) distribution characterized by a mean and standard deviation.
- c) Allowing a fraction of the containers to fail on emplacement. Experience has indicated that often a small fraction of the containers fail either due to emplacement practices or soon after emplacement. The improved models in DUST-MS permit the user to specify an initial failure fraction while allowing the remainder to fail based on the selected distribution and input parameters.

This paper presents the improved container failure models and discusses their implementation in DUST-MS. The distribution in container failure times requires that wasteform release calculations be calculated using a convolution integral. The approach used in DUST-MS to accomplish this is also presented. Extensive verification tests were performed covering all four leaching models in DUST-MS (rinse, diffusion, uniform, and solubility limited) and the effects of ingrowth in the wasteform prior to release. Results of the verification tests are presented. This is followed by a discussion of the importance of distributed failure on release and performance assessment. Finally, an example of the effects of failure rate is presented for a two-species decay chain in which the first species has a half-life less than the mean container failure time.

CONTAINER FAILURE MODELS

DUST-MS is a one-dimensional (1-D) model that predicts the release and transport of contaminants disposed in the subsurface. The conceptual model collapses the 3-D physical system down to 1-D mathematical representation. This implies that there are frequently multiple containers represented in one computational cell by a single effective container. This effective container can fail at a specified time that represents the mean time to failure of all containers represented in the computational volume. However, in practice it is probable that the containers will fail over a distribution of times. To account for this, the single failure time is generalized to a distribution of failure times. In theory, the distribution can be any function. In most cases, the distribution of failure time approximates known statistical distribution functions such as the uniform, normal, lognormal, or exponential distribution functions. In DUST-MS three failure distributions are permitted: instantaneous, uniform, or normal.

- a) Instantaneous failure of all containers at time t_j

The failure distribution function, which represents the rate of change in container failures as a function of time, is:

$$f(t - t_j) = \delta(t - t_j) \quad (1)$$

where $\delta(t - t_j)$ is the Kronicker delta function. The only information required for this model is the time to failure.

This is the release rate for a single failure time for all containers and is the model in the previous versions of DUST-MS.

- b) Uniform Container Failure Rate

The containers fail at a uniform rate from the beginning time of failure, t_b , to the ending time of failures, t_e . The failure distribution function is:

$$\begin{aligned} f(t - t_j) &= 0 & t < t_b \\ &= \frac{1}{(t_e - t_b)} & t_b \leq t \leq t_e \\ &= 0 & t > t_e \end{aligned} \quad (2)$$

The information required for this model is the failure start time, t_b and the failure finishing time, t_e .

- c) Normal distribution failure rate.

If the container failure rate follows a normal distribution, the distribution function is:

$$f(t) = \frac{1}{\sigma \sqrt{2\pi}} e^{-(t - \mu)^2 / (2\sigma^2)} \quad (3)$$

where σ is the standard deviation and μ is the mean value for failure times.

WASTEFORM RELEASE CALCULATIONS

With a distribution of failure times, calculation of release from the wasteform becomes more complicated than for a single failure time. To calculate release with a distribution of failures requires the combination of the fraction of containers failed at a given time and the release rate over the time since container failure. This can be represented as a sum over all containers:

$$R(t) = \sum_{j=1}^n r_j(t - t_j) f(t_j) \quad (4)$$

where $R(t)$ is the total release rate from all waste packages

$r_j(t - t_j)$ is the release rate from waste package j at time $t - t_j$, $t > t_j$

t_j is the failure time of the j^{th} container, and

$f(t_j)$ is the fractional rate of containers that fail at time t_j (in statistics this is known as the probability density function).

As an example, consider three containers with three different failure times, (10, 20, and 30 years). The total release rate from all three after 15 years would be:

$$R(15) = 1/3 r_1(t - \tau_1) = r_1(5)/3$$

Containers 2 and 3 have not failed at this time. Note this approach assumes that the mass is distributed uniformly between the three containers. Therefore, the total release is scaled by the factor of 1/3 which represents the fraction of containers that fail at a given failure time. After 40 years, the release would be:

$$R(40) = 1/3 r_1(30) + 1/3 r_2(20) + 1/3 r_3(10)$$

The above approach is appropriate when modeling only a few containers with known failure times. However, when attempting to model a large number of containers it becomes more computationally efficient to represent the failure times with a continuous distribution that represents the range of possible failure times. With a continuous distribution of failure times, Eqn (4) can be generalized as a convolution integral:

$$R(t) = \int_0^t r(\tau) \cdot f(t - \tau) d\tau \quad (5)$$

In Equation (5), we have assumed that all containers represented by the distribution of failure times have identical release rates. That is, one set of release rate parameters describes all of the containers in this computational volume. This is less general than the example given for the discrete case where each container was allowed to have unique release properties. However, it is required due to the impracticality of defining a unique set of release parameters that varies as a function of failure time. Equation (5) assumes that the failure rate and release rates are independent processes. This is true for a single radionuclide, but is not necessarily true for species in a decay chain. In that case, the release rate depends on the time for ingrowth and decay. Approximate methods are used to calculate release in this situation.

SPECIAL CASES OF WASTE FORM RELEASE RATES

a) Instantaneous failure of all containers at time t_j

Use of the instantaneous failure distribution, Eqn (1), in Eqn (5) gives the release rate as:

$$\begin{aligned} R(t) &= 0 & t < t_j \\ R(t) &= r(t - t_j) & t > t_j \end{aligned} \quad (6)$$

Where t_j is the time of failure for container j . This is the release rate for a single failure time for all containers in the control volume and corresponds to the model previously in DUST-MS.

b) Uniform Container Failure Rate

The containers fail at a uniform rate from the beginning time of failure, t_b , to the ending time of failures, t_e .

Using the uniform failure rate, Eqn (2), in Eqn (5) gives:

$$\begin{aligned}
 R(t) &= 0 & t < t_b \\
 &= \frac{1}{(t_e - t_b)} \int_{t_b}^t r(t - \mathbf{t}) d\mathbf{t} & t_b \leq t \leq t_e \\
 &= \frac{1}{(t_e - t_b)} \int_{t_e}^t r(t - \mathbf{t}) d\mathbf{t} & t > t_e
 \end{aligned} \tag{7}$$

c) Normal distribution failure rate.

Using this distribution, the release rate becomes:

$$R(t) = \frac{1}{\sqrt{2p}} \int_0^t \frac{r(t - \mathbf{t})}{s} e^{-(t - \mathbf{m})^2 / (2s^2)} d\mathbf{t} \tag{8}$$

Introducing a change of variables, $\tau' = (\tau - \mu)/\sigma$ leads to the following expression for release rate.

$$R(t) = \frac{1}{\sqrt{2p}} \int_{-m/s}^{(t-m)/s} r(t - s\mathbf{t}' - m) e^{-\mathbf{t}'^2 / 2} d\mathbf{t}' \tag{9}$$

FAILURE AT EMPLACEMENT

The user may specify failure of a fraction of the containers at emplacement. When this occurs, the release rate equations, (6) – (9), are modified by multiplication by the term $(1 - I_f)$ where I_f is the fraction of containers failed at emplacement. To account for the initial failures, another term is added to the release rate equation as follows.

$$R(t) = I_f r(t) \tag{10}$$

For the normal distribution, when the standard deviation is close to the mean, the model may predict a large fraction of containers to fail upon emplacement. For example, assume a mean value of 40 years and a standard deviation of 20 years. In this case, the distribution would predict 2.2% of the containers fail before time, $t = 0$. Thus integration from $t=0$ out to $t = \infty$ would predict failure of only 97.8% of the containers. Within DUST-MS, this is treated by declaring that 2.2% of the containers fail at emplacement.

RELEASE RATE MODELS

In DUST-MS, release from a wasteform is governed by one of four processes; surface rinse, diffusion, dissolution, and solubility limited. The models for wasteform release in DUST-MS have been described in detail elsewhere (1,2). To solve for the wasteform release rate, the appropriate release rate equation, (Eqn 6 – 9), is solved numerically. It should be noted that Equations (7) and (8) are convolution integrals that require a new integration at each computational time step. To enhance numerical efficiency, values of the probability density function and release rate are stored to prevent having to recalculate them.

VERIFICATION TESTS

Verification testing has been performed and involves comparison of code generated failure times with the input distribution and determination that leaching calculations are being performed correctly. Table I presents the major categories of test cases. In all, over 70 test cases were conducted. The complete details of the verification tests can be found in (3).

Table I: Matrix of Test Cases.

	Instant Failure	Gaussian Failure	Uniform Failure Rate
Rinse Release a) Solubility Limited b) Ingrowth* c) Initial Failure	Verified a-c.	Verified a-c.	Verified a-c
Diffusion Controlled Release d) Solubility Limited e) Ingrowth** f) Initial Failure	Verified d-f	Verified d-f	Verified d-f
Uniform Release g) Solubility Limited h) Ingrowth*** i) Initial Failure	Verified g-i	Verified g-i .	Verified g-i

* Ingrowth is calculated using the Bateman equations for all radionuclides prior to failure in the rinse model.

** Ingrowth model is exact only if all radionuclides have the same diffusion coefficient.

*** Ingrowth model is exact only if all radionuclides have the same fractional release rate.

Verification was tested through a number of methods. In the rinse model, the release is instant upon container failure. Therefore, for distributed container failure, it was verified that the release rate equaled the container failure rate specified through input for the problem. In addition, in the rinse model, when ingrowth occurs, it follows the decay equations known as the Bateman equations. In this case, release is the container failure rate multiplied by the inventory available at the failure time as specified by the Bateman equations. This was verified.

For the diffusion models, the instant failure model was verified through comparison to known analytical solutions for diffusion-controlled release. For the distributed failure models, there is no exact analytical solution. Therefore, two approaches were used to verify the models. In the first approach, the distribution of failure times was set to be extremely narrow so that it approximated an instant failure. For the Gaussian model, this is achieved by having a small (relative to a simulation time step), standard deviation. For the uniform model, this is achieved by having the end of the failures close to the start of the failures (less than one computational time step). In these cases, the distributed failure approaches an instant failure and the release rates from the distributed failure model were compared to the instant failure model. In all cases, agreement was within 1%. This provides confidence that the algorithms to calculate the convolution integral needed to estimate release with distributed failures is working. When the container failures are distributed over many time steps, approximate verification was achieved by comparing the release rate curves for a distributed failure with a mean failure time to release rates from an instant failure at the mean failure time. For long-lived radionuclides (i.e., decay is not important in the calculation), the total release at times long after container failures have been completed should be similar in the two cases. An example of this type of analysis is presented in the next section.

The ingrowth model for diffusion-controlled release in DUST-MS is exact when all species have the same diffusion coefficient. This was verified by simulating decay chains in which the last member has a long half-life as compared to the problem simulation time. Thus, the total release from all species in the chain, should equal the total release from a non-decaying species. This was verified for instantaneous and distributed container failure rates. When different species have different diffusion coefficients, the analytical models in DUST-MS for diffusion-controlled release are not accurate for ingrowth. Release is underpredicted if the progeny have a larger diffusion coefficient than the parent. The reverse is true for the progeny having a smaller diffusion coefficient than the parent. To alleviate this problem, DUST-MS allows the user to calculate diffusion release using a finite-difference model. This finite-difference model has been verified for ingrowth on analytical problems in which the diffusion coefficients are different (1). However, the finite-difference model has not been tested for distributed container failure models.

For the uniform release models, the instant failure model was verified against analytical solutions. An analytical solution is also available for the case of uniform container failure distribution and a uniform wastefrom release rate. The predictions of DUST-MS were compared to the analytical solution and agreement within 1% on predicted release was obtained. As with the diffusion model, when ingrowth occurs, an exact solution is obtained only when the parent and progeny have the same uniform release rate. The model was verified to be correct under this condition.

For all models, tests were conducted to demonstrate the solubility limited release model. In all cases, when the release rate was high enough to cause the solution concentration to exceed the solubility limit, the release rate is reduced to maintain solubility. This was demonstrated with all combinations of release models and container failure distributions.

ILLUSTRATIVE TEST PROBLEM

To demonstrate the effects of failure distribution on release, a test problem considering diffusion-controlled release from a cylindrical wastefrom is presented. In this problem, the first species, Species A, has a 44.7 year half-life, while the second species, Species B, has a 7.7×10^4 year half-life. Three container failure modes are modeled, instant at 100 years after emplacement, Gaussian with a mean life of 100 years and a standard deviation of 25 years, and uniform failure rate starting at 50 years and ending 150 years after emplacement. The disposal facility was started in 1950, however, this group of containers was not buried until 1965. Thus, the instant failure time is at 115 years from the problem start time, 1950. The waste form originally contains 1 gm of Species A and Species B is absent. Species B is produced from the radioactive decay of Species A. The wastefrom has a radius of 25 cm and both species have a

diffusion coefficient of 10^{-8} cm²/s. These values cause release to occur over hundreds of years (much longer than the range in container failure times). Transport away from the wasteform after release is controlled by advection with a Darcy velocity of 10^{-6} cm/s (31.5 cm/yr).

Figures 1 and 2 demonstrate the total release of Species A and B as a function of time and container failure rate. For Species A, Figure 1, the instant failure model begins release 115 years after the problem start time and has the lowest total release. This is due to the releases starting later and the effects of radioactive decay. The total release is less than 0.07 grams from an initial inventory of 1.0 grams. The release for the Gaussian failure model begins at the earliest of the three models due to the tails of this distribution. The uniform failure model releases the most of Species A, approximately 0.075 grams. All of Species A that decays prior to release is converted to Species B. The release of Species B is displayed in Figure 2. In this case, the instant failure model has the highest total release of Species B, releasing 0.54 grams. The cumulative release at the end of the problem simulation time is within 1% for all three container failure models. Thus, in terms of total mass release, the result is insensitive to container failure rate.

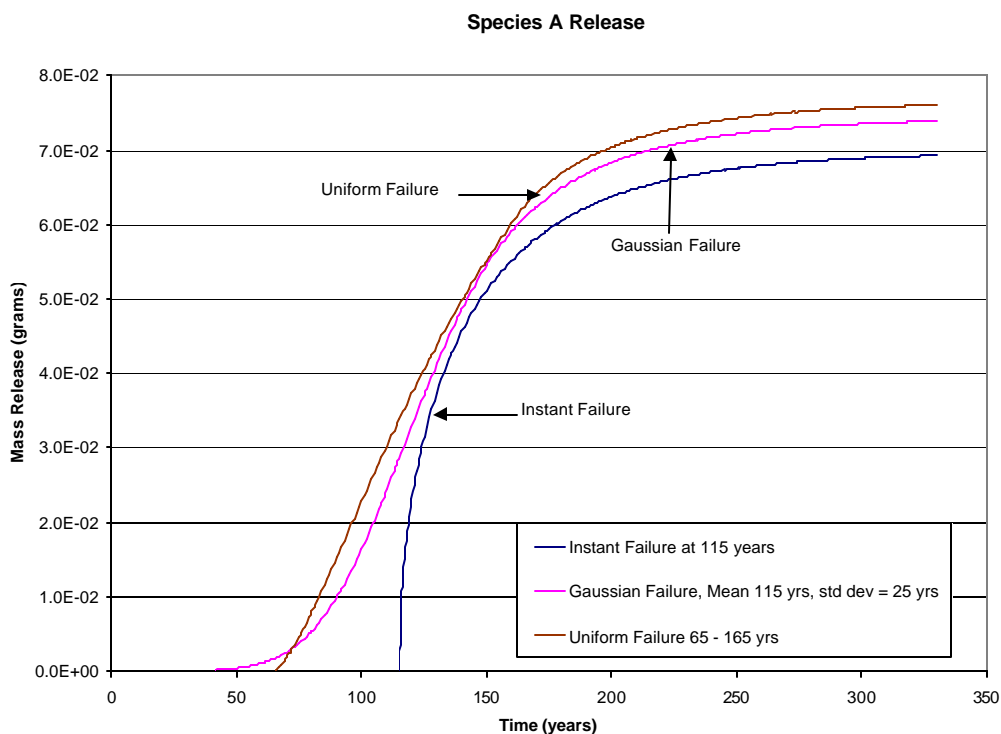


Fig. 1. Mass Release for Species A as a function of container failure rates.

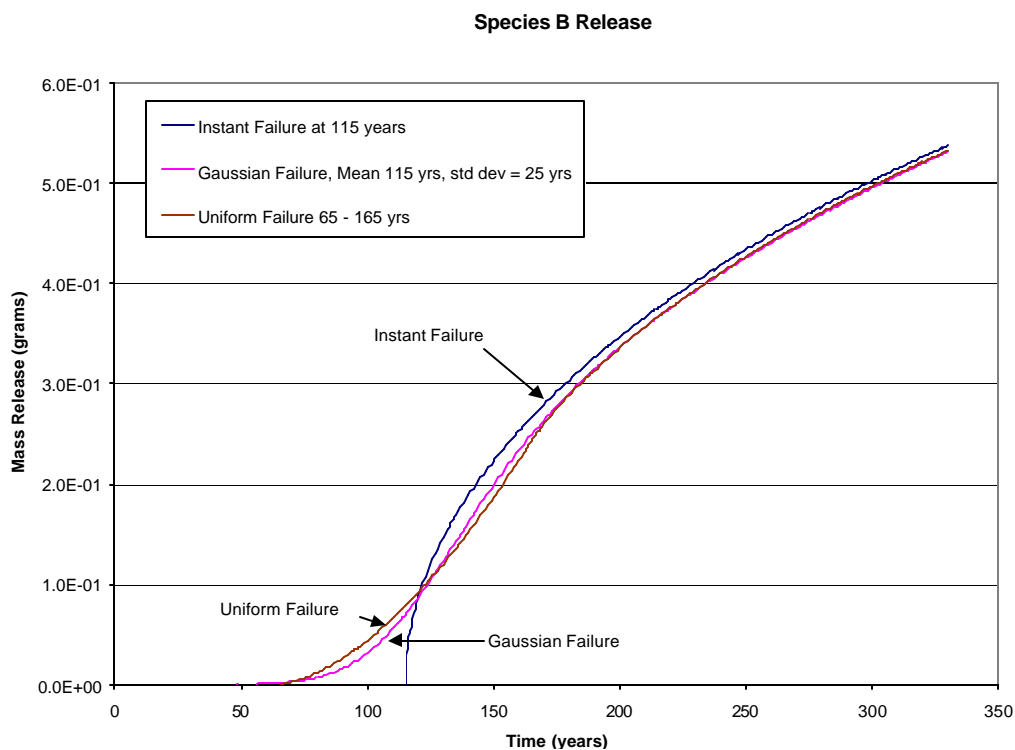


Fig. 2. Mass Release for Species B as a function of container failure rates.

Figures 3 and 4 display the concentration in the solution contacting the wasteform as a function of time. As material is released from the wasteform, the concentration increases. It decreases in time due to radioactive decay and advection moving the radionuclides away from the wasteform. In both cases, Figures 3 and 4, the instantaneous failure model has the highest solution concentration. This is expected because in this model, all containers fail at a single time. For Species A, the peak concentration is $2 \cdot 10^{-9} \text{ g/cm}^3$ when instantaneous failure occurs. For the Gaussian and uniform failure rates, the peak concentration is less than $3 \cdot 10^{-10} \text{ g/cm}^3$, almost an order of magnitude lower than the instantaneous case. For Species B, Figure 4, similar results occur. For the instantaneous failure model, the peak concentration is $7.1 \cdot 10^{-9} \text{ g/cm}^3$. For the Gaussian and uniform failure rates, the peak concentration is less than $1.3 \cdot 10^{-9} \text{ g/cm}^3$. For the uniform failure rate, the peak in Species A concentration occurs approximately between 90 and 100 years. Radioactive decay causes the concentration to decrease after this time even though additional containers fail until 165 years. For Species B and a uniform container failure rate, the concentration increases until 165 years when all containers have failed. After this time, the concentration decreases due to transport away from the waste form. For the Gaussian failure rates, the concentration follows a smoother distribution reflecting the container failure rate.

The difference in predicted concentration as a function of the failure model is a function of many parameters including radioactive decay, container failure rate, wasteform release rate, and the parameters that define transport away from the wasteform. However, as shown in this example, it may be substantial. Considering that most performance objectives for a waste disposal facility are concentration or dose (which is linearly proportional to concentration) based, the use of distributed failure models can have a significant impact when assessing performance.

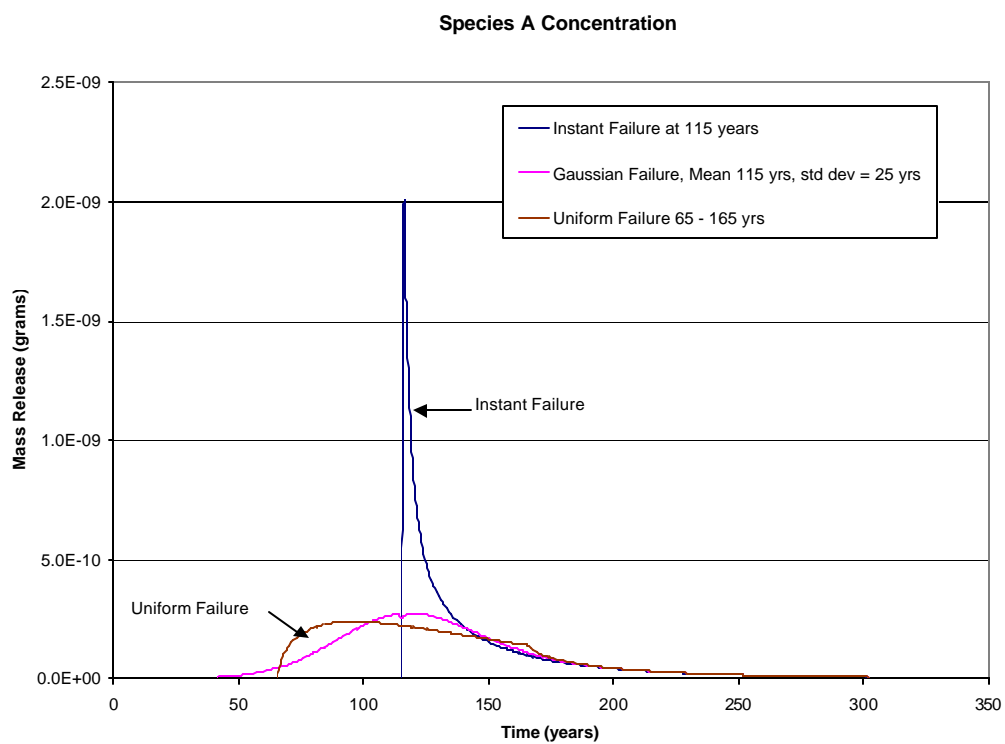


Fig. 3. Species A concentration as a function of container failure rate.

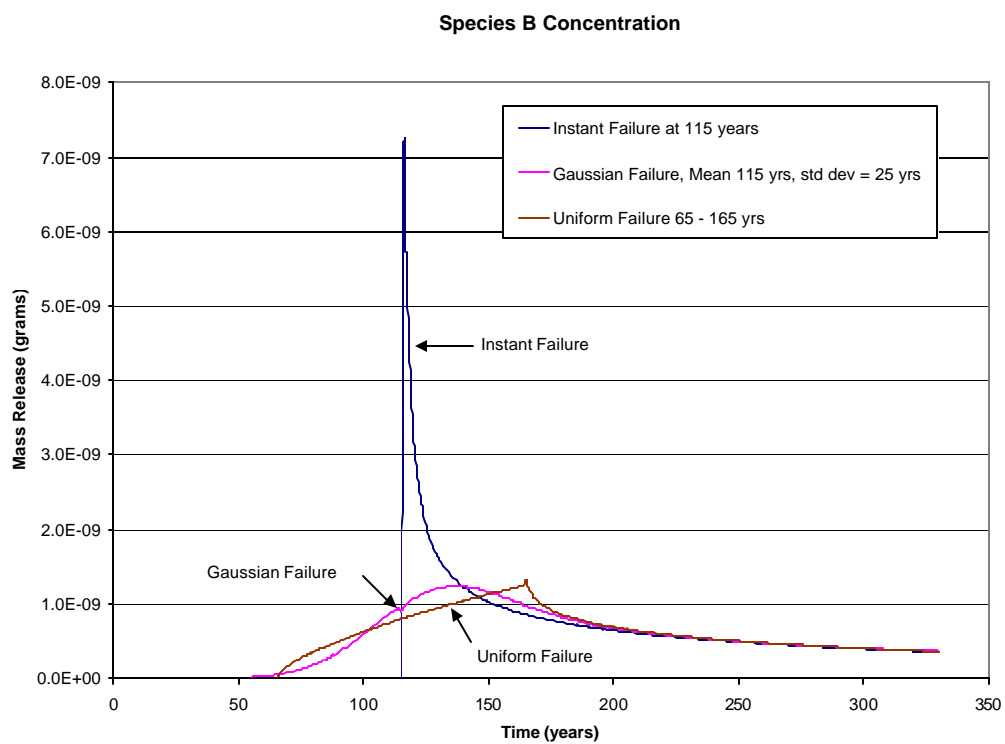


Fig. 4. Species B concentration as a function of container failure rate.

CONCLUSIONS

New models have been added to the DUST-MS code. These models permit simulating distributed failure (instantaneous, uniform or normal failure rate) of containers over time. Containers can be placed in the facility at different times to simulate disposal over the facility lifetime. In addition, a fraction of containers can be modeled as having failed at emplacement. These models have been implemented in the DUST-MS code and verified. An example calculation demonstrated that the failure rate had only a minor impact on total mass released, but had a more significant impact on the peak solution concentration. Simulating distributed failures lowers the predicted peak concentration. Peak concentration is often an important performance measure for a disposal facility.

ACKNOWLEDGEMENTS

This work was performed under the auspices of the U.S. Department of Energy, and was supported through Idaho National Engineering and Environmental Laboratory under the direction of Bruce Becker and Paul Ritter.

REFERENCES

1. Sullivan, T.M., "DUST-MS – Disposal Unit Source Term – Multiple Species", Informal Report, BNL-EWMG, 1998.
2. Sullivan, T.M., "**DUST** - Disposal Unit Source Term: Data Input Guide." NUREG/CR-6041, BNL-NUREG-52375, 1993.
3. Sullivan, T.M., "DUSTMS-D, Disposal Unit Source Term, Multiple Species, Distributed Container Failure, Data Input Guide," In preparation, January, 2001.